

REMARKS

By this amendment, applicants have amended claim 1 to delete the limitation that the voidage of the secondary particle is 2.5 to 35% and have recited this limitation in new dependent claim 18. Claim 1 has been amended to recite that the Ni content is $0.33 \leq y < 0.5$. The same nickel content is recited in claims 10 and 15. This amendment is supported by Figure 7 in which the Ni content (x) is from about 0.33. Claim 10 has been amended to be independent form and to further recite that the lithium secondary battery is for an automobile. See, e.g., page 1, lines 8-24 and the paragraph bridging pages 2 and 3 of Applicants' specification. Claim 15 is similar to claim 11 but, inter alia, recites that the length in which the plurality of primary particles are linked on a section of the secondary particle through a substantial center of the secondary particle is equivalent to 50 to 70% of the length of the whole periphery of the plurality of primary particles on the section of the secondary particle. See, the paragraph bridging pages 4 and 5 and the paragraph bridging pages 10 and 11 of Applicants' specification. Claim 11 has been canceled without prejudice or disclaimer, claims 12 and 14 amended to depend from and be consistent with claim 10, and new dependent claim 16-18 added to define further aspects of the present invention.

Claims 1, 6 and 10-14 stand rejected under 35 U.S.C. 103(a) as being unpatentable over Japanese Patent Application Publication Number JP-2001-243951 (Matsumoto et al.) in view of International Application Publication Number WO 03/044881 (Shiozaki et al.). Applicants traverse this rejection and request reconsideration thereof.

One feature of the present invention is in that the length in which the plural primary particles are linked on the section of the secondary particle is equivalent to

10 (preferably 50) to 70% of the length of the whole periphery on the section of the plural primary particles. This feature is based on the fact that the inventors of the present invention had found as described on page 4, lines 1-8 of the specification in English text. That is, the ionic conductivity is deteriorated at low temperature, since the viscosity of electrolytic solution decreases at low temperature. If each secondary particle of positive electrode material is formed with flocculated primary particles and the primary particles are positioned apart from one another so that the spaces among the primary particles are filled with electrolytic solution, a conductive network among the primary particles is locally decreased. Therefore, the resistance increases and the voltage drop of a battery occurs.

It is necessary to maintain the ionic conductivity to prevent the voltage drop of a battery. There are two ways to maintain the ionic conductivity, one being by maintaining the ionic conductivity of lithium in electrolytic solution, the other being by maintaining the diffusion of lithium ions between the primary particles.

The inventors of the present invention have found the fact that at low temperature, the diffusion of lithium ions between the primary particles is more important than the traveling of lithium ions via electrolytic solution, since the ionic conductivity is deteriorated at low temperature. Accordingly, by increasing the contact areas among primary particles, it is possible to maintain the conductive network in the environment of low temperature.

Fig. 2 shows that a secondary particle 2 of positive electrode material is formed with flocculated primary particles 1. For the cross section of each of the plural primary particles, the total length of the shared sides with the adjoining primary particles is calculated and compared with the length of the periphery of the primary particle. The desirable average of such contact length compared with the periphery of the section of the primary particle is 10 (preferably 50) to 70%.

Generally, volume change in a positive electrode material will occur when lithium ions come into it or go out from it due to charging/discharging. If the volume change in the positive electrode material is relatively large and the contact areas among primary particles are increased, primary particles may be crushed at the contact areas due to the volume change. This causes the life span of the battery to decrease.

Another feature of the present invention is in that the secondary particle is represented as $\text{Li}_a\text{Mn}_x\text{Ni}_y\text{Co}_z\text{O}_2$, and the secondary particle is composed of crystals having layer structure of composite oxide meeting $1 < a \leq 1.2$, $0 \leq x \leq 0.65$, $0.33 \leq y < 0.5$, $0 \leq z \leq 0.65$ and $x + y + z = 1$. In other words, the second feature of the present invention is in that the percentage content of Ni (y) is less than 50%, so that the relative lattice volume change rate decreases (on page 15, lines 23-24).

Fig. 7 shows that if the percentage content of Ni is 50% or less, the change of lattice volume due to charging/discharging decreases. Therefore, it is possible to maintain the high contact areas among the primary particles, and the high capacity maintenance rate.

As described above, there is a trade-off relation between the low temperature performance and the life span of the battery. If the low temperature performance is to be improved, the contact area among the primary particles should be increased to promote the diffusion of lithium ions between primary particles, which causes the life span of battery to decrease. If the life span of battery is to be increased, the contact areas among the primary particles should be decreased to prevent the primary particles from being crushed, but the low temperature performance is deteriorated.

In order to maintain both the low temperature performance and life span of battery, it is necessary to avoid secondary particles that include only such primary particles that the contact areas among the primary particles are small.

By combining these features, a lithium secondary battery for an automobile using the positive electrode material in which the discharge rate characteristic and the battery capacity in the environment of the low temperature of -30°C are hardly deteriorated and which are excellent in a cycle characteristic can be achieved.

Japanese Patent Publication 2001-243951 (Matsumoto et al.) discloses a positive electrode active material for a non-aqueous electrolyte secondary battery, which is comprised of secondary particles, each of secondary particle being composed of fine primary particle of lithium cobalt oxide. At least a part of the fine primary particles in a secondary particle are arranged in radiation toward outside from the center of the secondary particles. The secondary particles have many fine gaps among primary particles, in which the electrolyte can infiltrate.

Since the primary particles of Matsumoto et al., are arranged in radial directions in the secondary particles, gaps among primary particles vary from the center of the secondary particle to the periphery portion.

At the center of the secondary particle, gaps among primary particles are relatively small, but, at the periphery portion of the secondary particle, gaps among primary particles are relatively large, as shown in the attached Sheet No. 3. Accordingly, since the contact areas among primary particles are relatively large at the center of the secondary particle of Matsumoto et al., the primary particles at the center of the secondary particle may be cracked due to volume change caused by incoming and outgoing of lithium ions during charge/discharge. Such crack of the primary particles at the center of the secondary particle decreases the life span of battery.

Since the contact areas among primary particles are relatively large at the periphery of the second particle of Matsumoto et al., the gaps among the primary

particles at the periphery of the secondary particle can maintain electrolyte. However, by sintering at least part of the primary particles, electric conductivity and particle density can be improved. In applicants' arguments filed June 9, 2008, applicants explained the gaps among primary particles at the center of the secondary particle of Matsumoto et al., and then explained the gaps at the periphery of the secondary particle. These explanations are not contradictory, as alleged the paragraph bridging pages 4 and 5 of the Office Action.

In the paragraph bridging pages 5 and 6 of the Office Action, it is alleged that the sintering temperature 900°C of Matsumoto is very close to the lower limit 950°C of the sintering temperature disclosed in Applicants' specification in connection with the manner in which the present invention is made. Further, it is contended that the results of the present invention have not been adequately demonstrated in comparison with the disclosure of Matsumoto et al.

Generally, however, a small difference of sintering temperature produces different products.

The attached Sheet No. 1 includes photos at different magnifications which show secondary particles sintered at 900°C, while the attached Sheet No. 2 includes photos at different magnifications which show secondary particles sintered at 975°C. The photos show that the temperature difference of 75°C produces different products, as shown particularly in photos at magnification of 20000.

The Comparative Example 1 described beginning at the last line of Applicants' specification discloses a positive electrode material burnt at 900°C. The specification describes that since burning temperature is low, the growth of the crystal is insufficient and there are few locations in which primary particles are in contact. Fig. 2 of Matsumoto shows that crystal growth is locally sufficient at the

center of the secondary particle, but the crystal growth at the periphery of the radial structure is not sufficient when produced with the low sintering temperature 900°C.

The purpose of the present invention is to solve the problem that diffusion of lithium ions is reduced in low temperature environments. Since lithium ions cannot diffuse sufficiently in electrolyte under low temperature, the electric conductivity among primary particles is reduced. This is a problem for a lithium secondary battery for an automobile. According to the present invention, the connection length of primary particles in secondary particle is 10-70% in order to solve such a problem. On the other hand, the positive electrode material can be deteriorated due to volume change of positive electrode material during charge/discharge. Such volume change of positive electrode material causes the cycle life span to be decreased. The inventors of the present invention recognized that since the volume change rate of Ni is relatively large, the content of Ni should be reduced to be less than 0.5%.

Matsumoto and Shiozaki fail to teach the performances of lithium ions in low temperature environments, for example, less than -30°C.

The Shiozaki et al. publication discloses a positive electrode active material for a lithium secondary cell having a high energy density and excellent in charging/discharging cycle performance, and a lithium secondary cell having a high energy density and excellent in charging/discharging cycle performance are disclosed. A positive electrode active material for a lithium secondary cell is characterized in that the composition of the active material is $\text{Li}_x\text{Mn}_a\text{Ni}_b\text{Co}_c\text{O}_2$ (where a, b, and c are values of a point (a, b, c) on a side of or inside a quadrilateral ABCD having vertexes A(0.5, 0.5, 0), B(0.55, 0.45, 0), C(0.55, 0.15, 0.30), and D(0.15, 0.15, 0.7) on a ternary state diagram showing the relationship among a, b, and c, and satisfy the expressions $0.95 < x/(a+b+c) < 1.35$) and in that the active material

contains a composite oxide having an α -NaFeO₂ structure. The lithium secondary cell comprises this active material.

By determining only the voidage of secondary particle, it is not possible to avoid primary particles which have small contact areas among primary particles. In manufacturing positive electrode material, such secondary particles which include primary particles having relatively small contact areas are inevitably produced.

To improve electron conductivity, it is necessary to avoid such primary particles having small contact areas.

According to the present invention, the length in which the primary particles are linked on the section of the secondary particle is equivalent to 10 (preferably 50) to 70% of the length of the whole periphery on the section of the primary particles and the percentage content of Ni (y) is less than 50%, so that the relative lattice volume change rate decreases.

By satisfying these requirements, a lithium secondary battery using the positive electrode material in which the discharge rate characteristic and the battery capacity in the environment of the low temperature of -30°C are hardly deteriorated and which are excellent in a cycle characteristic can be achieved. One skilled in the art could not have thought the present invention having those three features based on Matsumoto et al. and Shiozaki et al. The object of the present invention is different from that of Shiozaki et al. The Shiozaki et al. publication does not teach the contact area among primary particles in each secondary particle of the present invention.

For the foregoing reasons, the presently claimed invention is patentable over the proposed combination of Matsumoto et al. and Shiozaki et al.

In view of the foregoing amendments and remarks, favorable reconsideration and allowance of all of the claims now in the application are requested.

To the extent necessary, applicants petition for an extension of time under 37 CFR 1.136. Please charge any shortage in the fees due in connection with the filing of this paper, including extension of time fees, to the deposit account of Antonelli, Terry, Stout & Kraus, LLP, Deposit Account No. 01-2135 (Case: 1021.43559X00), and please credit any excess fees to such deposit account.

Respectfully submitted,

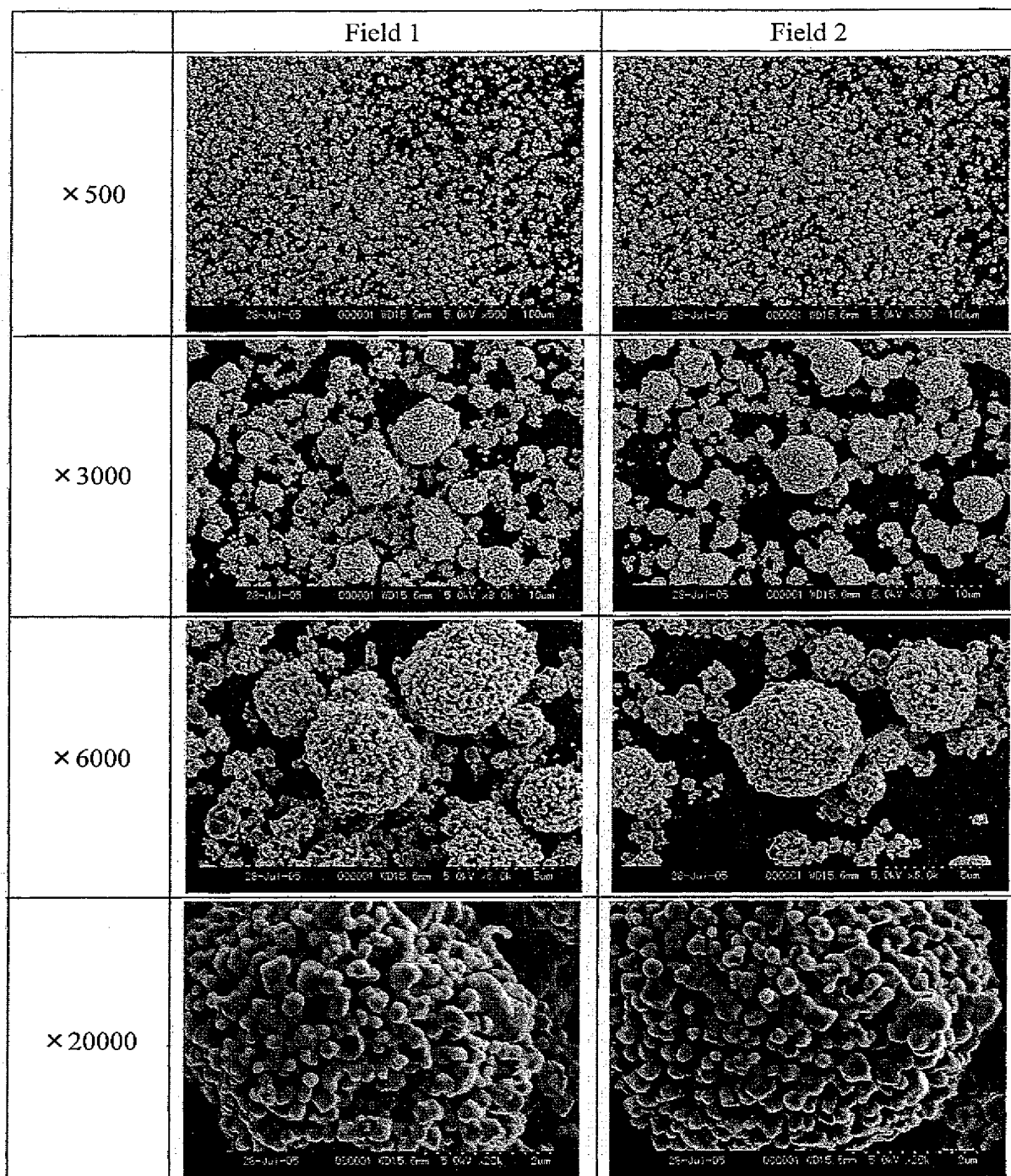
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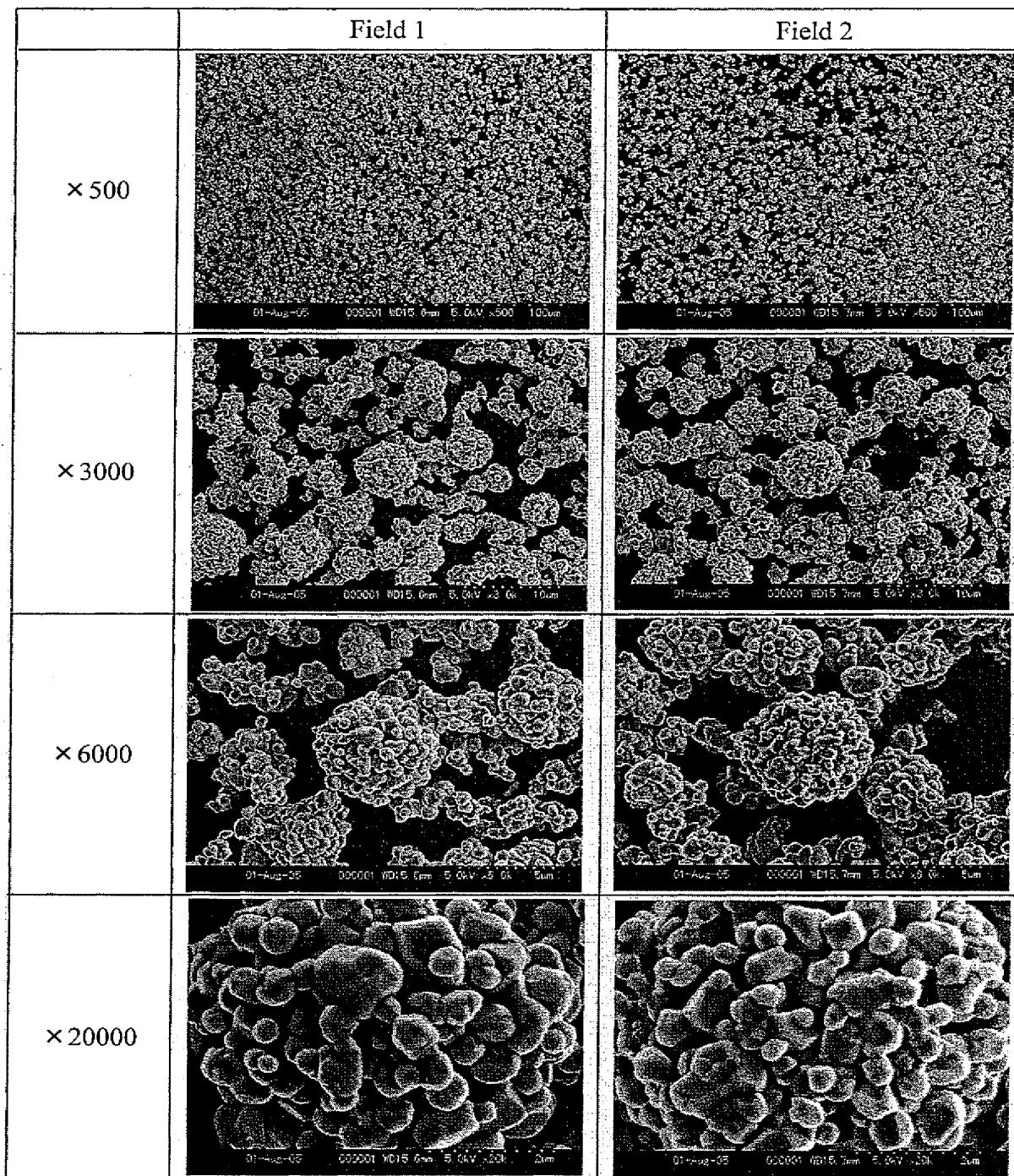
(BZ3-3)_Li1.0(High Viscosity)900°C_SEM Image of Granulated Powder



Powder=Active Material

05/07/28

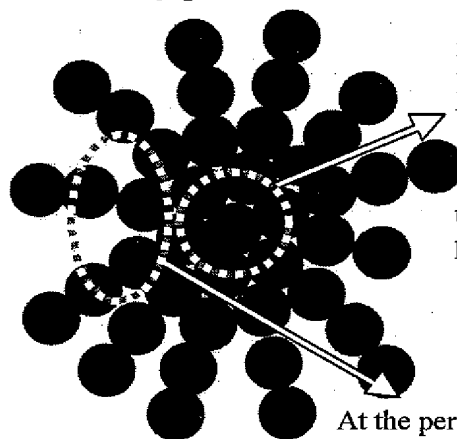
Li1.0(High Viscosity)975°C_SEM Image of Sintered Powder



Powder=Active Material

05/08/01

Secondary particle of JP'951

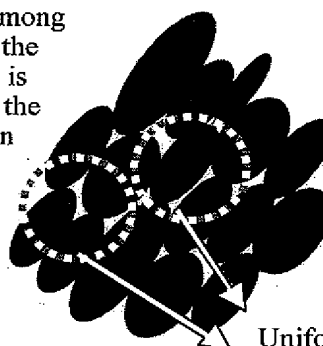


Radial Structure

At the center:
Length of Linkage among
primary particles on the
secondary particle is
more than 70% of the
whole periphery on
the section of the
primary particles.

At the periphery:
Length of Linkage among
primary particles on the
secondary particle is less
than 10% of the whole
periphery on the section
of the primary particles.

Secondary particle
of the present invention



Uniform Structure

Uniform:
Length of Linkage among primary
particles on the secondary particle is
10 to 70% of the whole periphery
on the section of the primary particles.